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Structural and optical properties of CdTe thin films deposited using RF magnetron sputtering

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Abstract

In this work, we have studied the influence of RF power on structural and optical properties of CdTe thin films deposited by indigenously designed locally fabricated RF magnetron sputtering. Films were analyzed by using variety of techniques such as low angle X- ray diffraction, UV-Visible spectroscopy, Raman spectroscopy etc. to study its structural and optical properties. Low angle XRD analysis showed that CdTe films are polycrystalline and has cubic structure with preferred orientation in (111) direction. Raman scattering studies revealed the presence of CdTe phase over the entire range of RF power studied. The UV-Visible spectroscopy analysis showed that the band gap decreases with increase in RF power. However, CdTe films deposited at higher RF power has optimum band gap values (1.44-1.60 eV). Such optimum band gap CdTe can be use as absorber material in CdS/CdTe and ZnO/CdTe solar cells.

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Keywords: CdTe; RF sputtering; Raman spectroscopy; Low angle XRD, UV-Visible spectroscopy

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1. Introduction

Cadmium Telluride (CdTe) is becoming the most successful contender to Si photoactive material for the realization of solar cells due to its high efficiency and low cost solar cell applications. The material is gaining great interest among the researchers due to its crucial properties such as long term performance stability [1], direct band gap (1.44 eV) that is near the optimum for photovoltaic solar energy conversion [2] and high optical absorption coefficient (> 10^5 cm^{-1}) to achieve a nearly full absorption of solar spectrum for thicknesses below 800 nm [3, 4].

Various deposition techniques can be used to the preparation of CdTe thin films includes closed space sublimation (CSS) [5], molecular beam epitaxy (MBE) [6], electrodeposition [7], pulsed laser deposition (PLD) [8, 9], metal organic chemical vapor deposition (MOCVD) [10], successive ionic layer adsorption and reaction method (SILAR) [11], screen printing [12], physical vapor deposition (PVD) [13], vacuum evaporation [14], electron beam evaporation [15], RF sputtering [16] and spray pyrolysis [17] etc. Among these methods RF magnetron sputtering method is most commonly used technique for the deposition of CdTe films and has been established for industrial applications. The RF magnetron sputtering permits deposition at low temperature, and gives better adhesion, larger coverage and higher film density than other methods. The main advantage of RF magnetron sputtering is that the stoichiometry of the sputtering material is retained in the deposited film making it a suitable technique for depositing intermetallic compounds [18]. Another advantage of the sputtering technique is the use of low energy particle bombardment for achieving lower growth temperatures along with the use of excited species for improving the doping control during growth [19].

The physical properties of CdTe films deposited by RF magnetron sputtering strongly dependent on process parameters such as sputtering power, argon gas pressure, substrate temperature, target-substrate distance etc. Enormous work has been done on structure, optical and electrical properties of CdTe thin films deposited by RF magnetron sputtering. However, still it requires further investigation to optimize the optical and structural properties to use as a suitable candidate for solar cell applications. With this motivation we have initiated the detailed study of synthesis and characterization of CdTe films using RF magnetron sputtering method. In this paper, we present the detail investigation of influence of RF power on structural and optical properties of CdTe films deposited by RF magnetron sputtering method.

2. Experimental

2.1. Film Preparation

The CdTe films were deposited on corning #7059 substrates using indigenously designed locally fabricated RF magnetron sputtering system [20]. Fig 1 shows the schematic diagram of RF magnetron sputtering system.

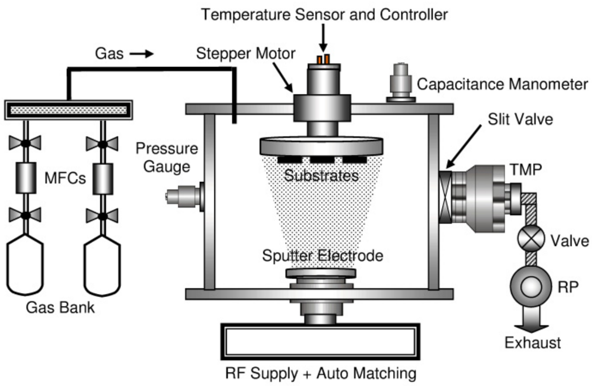

Fig. 1. Schematic diagram of indigenously designed locally fabricated RF magnetron sputtering system.

It consists of a cylindrical process chamber coupled with a TMP followed by a roughing pump which yields a base pressure less than 10^{-7} Torr. A target of 4 inch diameter (99.99 %, Vin Karola Instrument, USA) was used for deposition of CdTe films and was kept facing substrate holder ~ 7 cm away. To have uniform film substrates were kept rotating during the sputtering process using a stepper motor. The RF power was varied from 30 W to 150 W. The substrates can be clamped on substrate holder which is heated by inbuilt heater using thermocouple and temperature controller. The process pressure was kept constant by using throttle valve and measured with capacitance manometer. Argon (Ar) can be introduced in the process chamber through a specially designed gas bank assembly which consists of MFCs and gas mixing. Other process parameters employed during deposition of CdTe films are listed in Table 1.

Table 1. Process parameters employed during the deposition of CdTe films

Process parameter	Value
Process Pressure	1x10-2 mbar
Deposition Time	30 min
RF Power	30-150 W
Distance between substrate holder and target electrode	7 cm
Ar gas flow rate	60 sccm
Substrate temperature	100 0C

The substrates were cleaned using a standard cleaning procedure. Prior to each deposition, the substrate holder and deposition chamber were baked for two hours at 100 ^{0}C to remove any water vapor absorbed on the substrates and to reduce the oxygen contamination in the film. After that, the substrate temperature was brought to desired value by appropriately setting the inbuilt thermocouple and temperature controller. Sputter-etch of 10 min were used to remove the target surface contamination. The deposition was carried out for desired amount of time and films were allowed to cool to room temperature in vacuum.

2.2. Film characterization

Low angle X-ray diffraction pattern were obtained by X-ray diffractometer (Bruker D8 Advance, Germany) using CuKα line (λ = 1.54056 Å). The average crystallite size was estimated using the classical Scherrer's formula. The band gap of the films was deduced from transmittance and reflectance spectra of the films deposited on corning glass and were measured using a JASCO, V-670 UV-Visible spectrophotometer in the range 300-2500 nm by using the procedure followed by Tauc. Raman spectra were recorded with Raman spectroscopy (Jobin Yvon Horibra LABRAM-HR) in the range 50-1200 cm^{-1}. The spectrometer has backscattering geometry for detection of Raman spectrum with the resolution of 1 cm^{-1}. The excitation source was 532 nm line of He-Ne laser. The power of the Raman laser was kept less than 10 mW to avoid laser induced crystallization on the films. Thickness of films was determined by profilometer (KLA Tencor, P-16+) and was further confirmed by UV-Visible spectroscopy using the method proposed by *Swanepoel* [20].

3. Results and Discussion

3.1. Variation of film thickness

Variation of thickness of CdTe thin films as a function of RF power is shown in Fig. 2. As seen from the figure, the thickness of the film increases from 3.65 μm to 12.64 μm when RF power increased from 30 W to 150 W. In the sputtering process the growth rate of films mainly depends on the variation in sputtering process parameters such as sputtering gas, process pressure, target composition, operating power, substrate temperature and the distance between the target and the substrate etc. According to sputtering theory, the sputtering deposition rate of films is proportional to the product of the sputtering rate of the target and the density of the Ar ion stream. The density of the Ar ion stream increases with increase in sputtering power. As a result the sputtering rate of the target also increases. Consequently the growth rate of the film and hence thickness of the film increases with increase in RF power. The increase CdTe film thickness with increase in RF power was reported previously by *Guanghua et al.* [21].

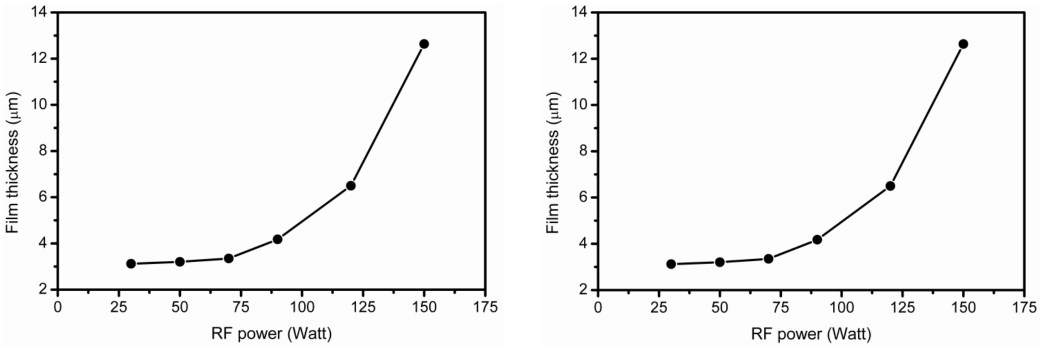

Fig 2. Variation of thickness of CdTe thin films as a function of RF power

3.2. Low angle x-ray diffraction analysis

Low angle x-ray diffraction (low angle-XRD) is a widely used as nondestructive technique for the structural characterization of different materials. Films deposited on glass were used for the low angle-XRD measurements. All low angle-XRD patterns were obtained at a grazing angle of 1^0. Fig. 3(a) display the low angle-XRD pattern of CdTe thin films deposited at different applied RF powers.

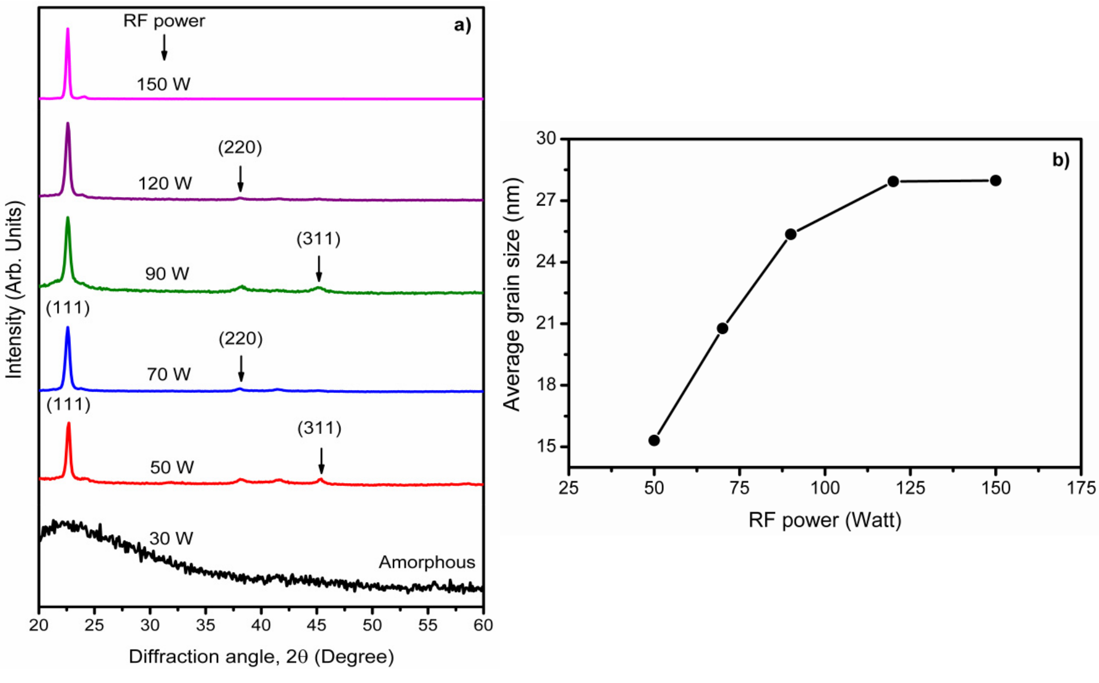

Fig. 3. a) Low angle-XRD pattern b) Estimated average grain size ($d_{x\text{-}ray}$) of CdTe thin films at different applied RF powers.

As seen from the XRD pattern the films deposited at low RF power (> 30 W) didn't show any diffraction peaks suggesting that the films are amorphous. However films deposited at high RF power (> 30 W) show three peaks at 2θ ~ 23.48^0, 38.50^0 and 46.17^0 corresponding to (111), (220) and (311) diffraction planes respectively which are characteristic of polycrystalline cubic structure of CdTe [JCPDS data card #15-0770]. The XRD pattern of the films deposited in the present study is consistent with the reported in the literature [22, 23] which confirm the formation of CdTe films. The dominant diffraction peak is (111) signifying that the films have preferred orientation in (111) direction. With increase in RF power the intensity of (111) diffraction peak increases signifying increase in crystallinity in the CdTe films. No other diffraction peaks due to impurities or phase were observed on XRD patterns, suggesting that the as-synthesized CdTe films were relatively pure.

The average grain ($d_{x\text{-}ray}$) size has been estimated using the classical *Scherrer's* formula [24],

$$d_{x\text{-}ray} = \frac{0.9\,\lambda}{\beta \cos\theta_\beta} \qquad .....(1)$$

Where λ is the wavelength of the x-ray used, β is full-width at half-maximum (FWHM) and θ is the Bragg's diffraction angle. The estimated average grain size ($d_{x\text{-}ray}$) as a function of RF power is shown in Fig. 3 (b). As seen the average grain size of CdTe films increases from 15.3 nm to 27.9 nm when RF power increased from 50 W to 120 W. However, further increase in RF power to 150 W there is no significant change in average grain size and it remains almost constant. These results suggest that 120 W is an optimized RF power of our RF sputtering unit to obtain CdTe thin films with moderate crystallinity and crystallite size.

3.3. Raman spectroscopy analysis

Raman spectroscopy is a very powerful non-destructive technique used to investigate the structure of materials because it gives a fast and simple way to determine the phase of the material, whether it is amorphous, crystalline or nanocrystalline. Fig. 4 shows Raman spectra of CdTe films deposited at different RF power using RF magnetron sputtering.

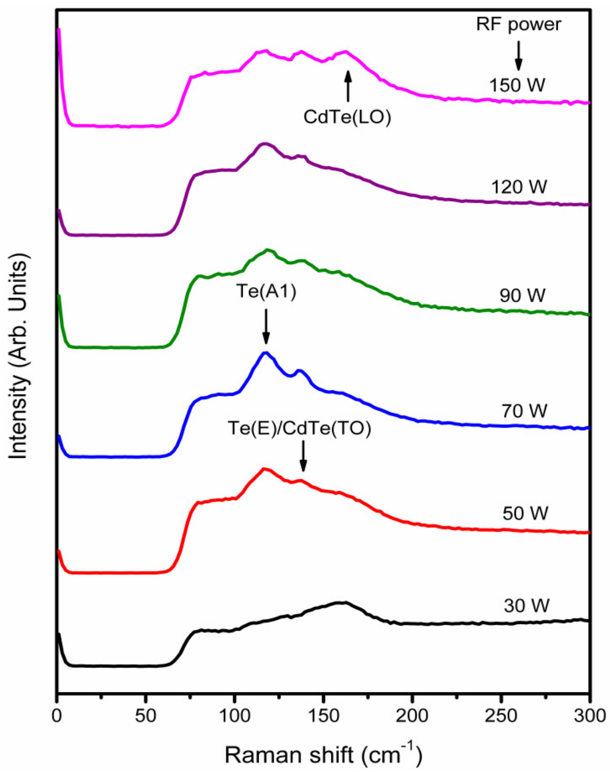

Fig. 4. Raman spectra of CdTe films deposited at different RF power using RF sputtering method.

As seen from the fig., the Raman spectra shows major Raman peaks centered ~ 162.7 cm^{-1}, 137.6 cm^{-1} and 116.8 cm^{-1}. The peak centered ~ 162.7 cm^{-1} corresponds to fundamental longitudinal optic (1 LO) phonon modes of CdTe, [25, 26]. The peak located ~ 137.6 cm^{-1} is overlapping of TO phonon in CdTe and E symmetry of Te and the peak centered ~ 116.8 cm^{-1} corresponds to trigonal Te [27, 28].which is overlap with each other. We do not observed bands around 450 $cm^{-}1$ in any sample (not shown), which have been related to bending vibrations of the Te-O-Te linkages in a continuous TeO_2 network [29-31]. Furthermore, no other Raman peak was observed over the entire range of RF power studied suggesting formation of single phase CdTe films. These results are consistent with low angle XRD analysis and further confirm the formation of CdTe films by RF sputtering method.

3.4. UV-Visible spectroscopy analysis

Optical properties of CdTe films have been deduced from transmission (T) and reflection (R) spectra of the films deposited on corning glass using UV-Visible spectrophotometer. Fig. 5(a) displays the transmittance curves of CdTe thin films prepared at different RF powers in the range 700-1100 nm.

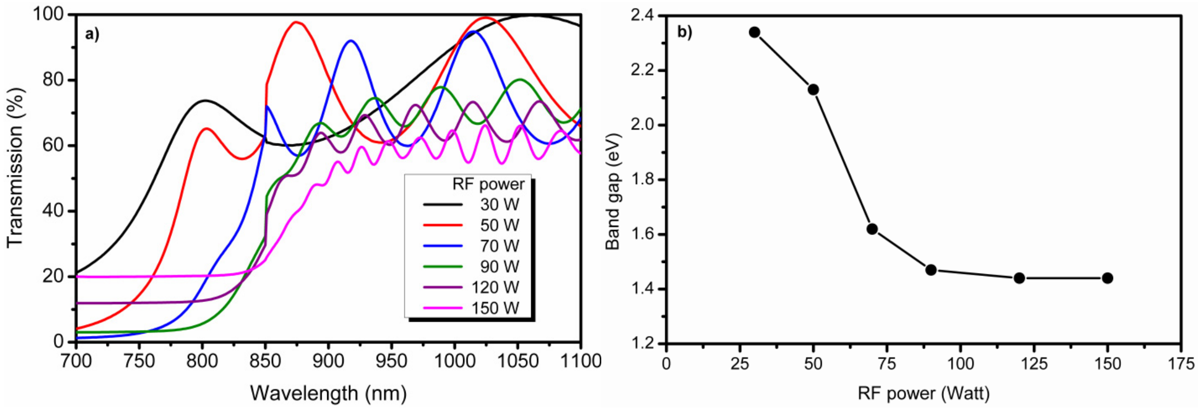

Fig. 5. a) Transmittance spectra b) Variation of optical band gap of CdTe thin films prepared at different RF powers by sputtering method

The presence of interferences fringes in films transmittance spectra indicates that films have smooth surface morphology. The transmission strongly depends on the film structure, which is determined by the preparation methods, film thickness and deposition conditions [32]. As seen from figure the average transmission was found > 60 % which is good for opto-electronic devices, especially for solar cell window layers. The higher transmittance can be attributed to less scattering effects, structural homogeneity and better crystallinity [33]. Furthermore, sharp fall at band edge is an indication of good crystallinity of CdTe in the films. The XRD analysis further supports this.

As CdTe is a direct band gap semiconductor [34, 35], the optical energy band gap (E_{opt}) and the optical absorption coefficient (α) are related by [36],

$$(\alpha E)^{1/2} = B^{1/2}(E - E_{opt}) \qquad ....(2)$$

Where α is the absorption coefficient, B is the optical density of state and E is the photon energy. The absorption coefficient (α) can be calculated from the transmittance of the films with the formula,

$$\alpha = \frac{1}{d} \ln\left(\frac{1}{T}\right) \qquad .....(3)$$

Where d is the thickness of the films and T is the transmittance. Therefore, band gap can be obtained by extrapolating tangential line to photon energy (E = hυ) axis in the plot of (αhυ) as a function of hυ. The variation of band gap as a function of RF power for CdTe thin films deposited using RF sputtering is shown in Fig. 5(b). As seen the optical band gap decreases from 2.34 eV to 1.44 eV when the RF power increased from 30 W to 120 W. It is interesting to note that the values band gap of CdTe films deposited at higher RF power (70 W) are in the range 1.44-1.60 eV which is in excellent agreement with the reported bulk CdTe band gap value [37-39] and support to the formation of stoichiometric CdTe films at higher RF power. These optical band gap values are in good agreement with the values reported by other authors. For example, *Ikhmayies et al.* [40] reported optical band gap of 1.48 eV value for vacuum evaporated CdTe films. *Choi et al.* [41], estimated the optical band gap energy from 1.41 to 1.45 eV for sputtered CdTe films. *Pandey et al.* [9] reported values of optical band gap of 1.6 eV value for cubic CdTe films and 1.54 eV for hexagonal CdTe films deposited by pulsed laser ablation. Recently, *E. Espinosa et al.* [23] have also obtained optical band gap values in the range 1.44-1.60 eV for CdTe films deposited by sputter technique.

4. conclusion

Thin films of CdTe were grown by RF magnetron sputtering at various RF powers. Structural and optical properties of these films were investigated by using various characterization techniques. Low angle XRD analysis showed that CdTe films are cubic polycrystalline and has preferred orientation in (111) direction. Furthermore, increase in RF power improves crystallinity in the CdTe films. Raman scattering studies revealed the presence of CdTe phase over the entire range of RF power studied. The UV-Visible spectroscopy analysis showed that the optical band gap decreases from 2.34 eV to 1.44 eV when the RF power increased from 30 W to 120 W. However, band gap of CdTe films deposited at higher RF power (> 70 W) are in the range 1.44-1.60 eV which is in excellent agreement with the optimum band gap value. Such optimum band gap CdTe can be use as absorber material in photovoltaic applications like the CdS/CdTe and ZnO/CdTe solar cells.

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